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(54) Name of invention

Resin compound and its' production method

(57) (Summary)

(Issue) To put forward a resin compound that has excellent impact resistance properties and at the same time, flow properties.

(Resolving procedure) A resin compound composed from 45-95% part weight of poly propylene, 55 - 5 % part weight of poly phenylene ether, a specific hydrogenated block

copolymer 1 – 30 % part weight, a chemical compound of 0.1 – 5 % part weight with specific active groups and 3 – 100 % part weight of inorganic filler.

(Scope of patent claim)

(Claim item 1)

- | | | |
|-------|------------------------------|------------------------|
| (a) | Poly propylene related resin | 45~95 % of part weight |
| (b) | Poly phenylene related resin | 5~5 % of part weight |

A resin compound that has the properties of containing the following:-

A hydrogenated block copolymer made from the hydrogenation of the block copolymer made up from atleast 1 copolymer block B with the main part as conjugated diene compound having 2 - 90 % total amount of 3,4- vinyl bonds and 1,2- vinyl bonds of conjugated diene compound and atleast one polymer block A having the main part of vinyl aromatic group compound as (c) component with regard to a total of 100 w/t parts of the above - mentioned (a) component, (b) component

1 - 30 w/t parts

A compound having active groups within the (d) molecule, that can be chosen from carbon- carbon double bonds and carbonic acid groups, anhydride groups, epoxy groups, amino groups and hydroxyl groups, with regard to a total of 100 w/t parts of the above - mentioned (a) component, (b) component

0.1 - 5 w/t parts

with regard to a total of 100 w/t parts of the above - mentioned (a) component ~ (d) component,

(e) inorganic filler

3~100 w/t parts

(Claim item 2) A resin compound that is mentioned in the claim item 1 and having the property of the inorganic filler with the (e) component present in a fibrous and / or plate form.

(Claim item 3) The manufacture method of the resin compound that is mentioned in the claim item 1 and has the properties of being manufactured by dissolving and kneading the entire amount or a part of (c) component, (a) component and the total amount of (b) component, (d) component, following which the remaining amount of (a) component, (c) component and (e) component is supplied which is then further subjected to dissolving and kneading action.

(Detailed description of the invention)

(0001)

(Fields of industrial use)

The current invention is concerned with a resin compound having excellent oil resistance properties, chemical resistance properties, heat resistance properties, impact resistance properties, rigidity properties, molding processing properties that can be used in the electrical / electronic fields, automobile fields and other types of industrial material fields, moreover it deals with a resin compound with excellent durability properties (impact resistance properties) as a heat resistant material.

(0002)

(Existing technology / Prior art)

The poly phenylene ether resin is known as an engineering plastic that has excellent flame retardant properties, heat resistance properties, measurement stability properties, low water absorption properties and electric properties, but has the drawbacks of having poor formation processing properties and poor solubility flow properties as well as having poor solvent resistance properties and impact resistance properties. On one hand, the poly olefin resin is a low priced plastic with low specific gravity and has excellent chemical resistance properties, solvent resistance properties and formation processing properties, and is being used in fields such as those of automobile parts and electric / electronic device components and domestic electric products. At this juncture, the development was foreseen for a resin compound having excellent formation processing properties, heat resistance properties and flame retardant properties by mixing both the resins, correcting the drawbacks of both and can be expected to be an excellent resin material with wide applications in the fields of usage.

(0003) For this reason, various proposals were made regarding the poly olefin/ poly phenylene related polymer alloy. For instance, in the American Patent Number 3361851 detailed report, by blending the poly phenylene ether and poly olefin, it was proposed to improve the solvent resistance properties and the heat resistance properties, in the American Patent Number 3994856 detailed report, it was proposed to improve the impact resistance properties and the solvent resistance properties by blending the poly phenylene ether and poly phenylene ether and stearine related resin with a hydrogenated block copolymer, in the American Patent Number 4145377 detailed report, it was proposed to improve the impact resistance properties and the solvent resistance properties by blending the poly phenylene ether and the stearine related resin with a hydrogenated block copolymer and a prepared mixture made up of polyolefin/ hydrogenated block copolymer = 20~ 80 w/t parts / 80 ~ 20 w/t parts.

(0004)

Furthermore, in the American Patent Number 4166055 detailed report and in the American Patent 4239673 detailed report, it has been detailed that there is an improvement of the impact resistance properties by blending the poly phenylene ether with hydrogenated block copolymer and poly olefin. Subsequently, in the American Patent Number 4383082 detailed report and in the European Patent Application 1151712 detailed report, it has been detailed that there is an improvement of the impact resistance properties by blending the poly phenylene ether with hydrogenated block copolymer and poly olefin.

(0005)

Further, in the patent Application S63 - 113057, patent application S63 - 225642, American Patent Number 4863997 detailed report and the patent application H3 - 72512, patent application H4 - 183748, patent application H5 - 320471, a resin compound with excellent chemical resistance properties and processing properties was proposed by mixing a specific hydrogenated block copolymer for the reformulation of a resin compound composed of poly olefin resin and poly phenylene ether resin.

(0006)

Further, said applicant has proposed a resin compound with excellent solvent resistance properties and excellent phase solubility properties, rigidity properties and heat resistance properties of the specific hydrogenated block polymer and the poly phenylene ether and poly olefin in the Patent Number H2 - 225563, Patent Number H3 - 18505, Patent Number H5 - 70679, Patent Number H5 - 295184, Patent Number H6 - 9828, Patent Number H6 - 16924, Patent Number H6 - 57130, Patent Number H6 - 136202.

(0007)

Furthermore, in the Patents number H4 - 28739, number H4 - 28740, the manufacture method is suggested for a resin compound that has an excellent balance of mechanical properties and, especially of impact strength and rigidity properties, being made up of a rubber type polymer body and hydrogenated block copolymer body or poly olefin and poly phenylene ether. And in the Patent number H7 - 166026, the phase solubility property of poly phenylene ether related resin and poly olefin related is good, and the manufacture method is suggested for a resin compound having excellent mechanical properties, especially impact resistance properties. Therefore, with regard to higher heat resistance properties and the demand for measurement frequency, it is suggested in the Patents H7 - 90183, Patent 2000 - 119454 that atleast one type of inorganic filler in fibrous form, plate form and granular form should be added.

(0008)

(Issues that the invention tries to resolve)

The resin compound that is actually made from poly phenylene ether and poly phenylene ether and the aromatic group vinyl compound polymer body the rubber type polymer and hydrogenated block polymer and the olefin related resins, and a resin compound with thermal plasticity properties was suggested according to the above- mentioned advanced technology in Patent Application Number H7 - 90183, Patent 2000 - 119454. However, in the above - mentioned resin compound with thermal plasticity properties, there is a tendency of remarkable lowering of the impact resistance property along with the improvement of flow property, and inversely when the impact resistance property is improved, there is significant lowering in the flow property. It is an object of the current invention to provide a resin compound that has excellent impact resistance properties as well as flow properties, from the proposed above - mentioned resin compound with thermal plasticity properties.

(0009)

(Procedure for resolving the issue)

The current inventors, upon considering the present condition, reached this invention by discovering a resin compound with excellent impact resistance properties as well as flow properties, that had not been achieved with the existing technology, in a specific resin by progressing in diligent study and in order to increase to a high degree, flow properties and impact resistance properties in the resin compound that is made up from the inorganic filler and the hydrogenated block copolymer that is used as an additive, poly propylene related resin and poly phenylene ether related resin.

(0010)

In other words, the current invention is a resin compound made up from,

- | | |
|---------------------------------------|------------------------|
| 1. (a) Poly propylene related resin | 45~95 % of part weight |
| (b) Poly phenylene related resin | 55~5 % of part weight |

A hydrogenated block copolymer made from the hydrogenation of the block copolymer made up from atleast 1 copolymer block B with the main part as a conjugated diene compound having 2-90 % total amount of 3,4- vinyl bonds and 1,2- vinyl bonds of conjugated diene compound and atleast one polymer block A having the main part of vinyl aromatic compound as (c) component with regard to a total of 100 w/t parts of the above- mentioned (a) component, (b) component

1 - 30 w/t parts

A compound having active groups within the (d) molecule, that can be chosen from carbon- carbon double bonds and carbonic acid groups, anhydride groups, epoxy groups, amino groups and hydroxyl groups, with regard to a total of 100 w/t parts of the above - mentioned (a) component, (b) component

0.1 - 5 w/t parts

with regard to a total of 100 w/t parts of the above- mentioned (a) component ~ (d) component,

(e) inorganic filler

3~100 w/t parts

(0011)

2. The manufacture method proposed consists of dissolving and kneading the entire amount or a part of (c) component, (a) component and the total amount of (b) component, (d) component, following which the remaining amount of (a) component, (c) component and (e) component is supplied which is then further subjected to dissolving and kneading action.

(0012)

The poly propylene related resin used as the (a) component in the current invention, is a crystalline propylene homopolymer and, the propylene obtained after the second polymerization process of the crystalline propylene homopolymer obtained in the first process of polymerization, a crystalline propylene- ethylene block copolymer having a propylene- ethylene random copolymer portion that is obtained by copolymerizing

ethylene and / or atleast one other α -olefin (for example, butane- 1, hexane- 1 etc), moreover these crystalline propylene homopolymers can also be the mixture of crystalline propylene- ethylene copolymer. The poly propylene related resin is normally obtained by polymerizing under the presence of an alkyl aluminium compound and a halogenised titanium catalyst etc that is carried on substrates such as titanium trichloride catalyst and magnesium chloride, in the range of the polymerization temperature 0 ~100 ° C and in the range of polymerization pressure 3~ 100 atmospheric pressure.

(0013)

At this juncture, it is possible to add a chain transfer agent such as hydrogen etc., in order to adjust the molecular weight of the polymer and as the polymerization method, either of the methods of batch method or continuous method is possible and solution polymerization under the solvents of butane, pentane, hexane, heptane, octane, etc., or slurry polymerization, etc can be selected and moreover, methods such as mass polymerization in a monomer under the absence of a solvent or a vapor phase polymerization method in a monomer of gas form, can be applied. Moreover, in order to increase the polymerization activity of the isotacticity of the poly propylene obtained other than the above - mentioned polymerization catalysts, it can be used as an external donor component or the internal donor component in the compound with electric donor properties as the third component, and these compounds of electric donor property are well known and used.

(0014)

For example, the following can be used- ester compounds such as ϵ - caprolactone, methyl methacrylate, ethyl benzoate, methyl toluate, ester phosphites such as triphenyl phosphite, tributyl phosphite, phosphorous acid derivatives such as hexamethyl phosphoric triamide, alkoxy ester compounds , aromatic monocarboxylic esters and/ or aromatic alkyl alkoxy cyrans, aliphatic hydrocarbon alkoxy cyran, each type of ether compound, each type of alcohol and/ or each type of phenol.

(0015)

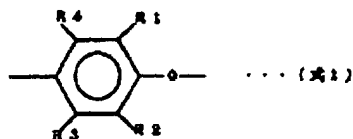
If the poly propylene related resins that are proposed in the current invention are obtained by the above mentioned methods, they can be used either singly or together at no matter what crystallization property and solubility points. Furthermore the melt flow rate (MFR) of the poly phenylene related resin (230 ° C, load 21.2N) is 0.01~ 300g/ 10 minutes, and preferably 0.1 ~100g/ 10 minutes and more preferably in the range of 0.3~ 50g/ 10 minutes. Besides, if it is the MFR of these ranges, it can be used either singly or together.

(0016)

Subsequently, the poly phenylene ether related resin (b) component of the current invention (abbreviated as PPE below), is the compound having the following depicted structure. The poly phenylene ether is a widely known compound by itself. Bond unit:

(0017)

(Formula 1)



.....(Formula 1)

(0018)

(Here, R1, R2, R3 and R4 are respectively hydrogen, halogen, first grade and second grade low grade alkyl groups of carbon number 1~7, phenyl groups, haloalkyl groups, amino alkyl groups, hydrocarbon oxy group and atleast 2 halo hydrocarbon oxy groups selected from the groups of halo hydrocarbon oxy group that separates the halogen atom and the oxygen atom, and can be different from each other) and is a homopolymer and/or copolymer with reduced degree of viscosity (0.5/dl, chloroform solution, 30 ° C measurement) within the range of 0.15 ~2.5, preferably 0.30~ 2.00 and more preferably in the range of 0.35 ~2.00.

(0019)

As examples of PPE, the following poly phenylene ether copolymers such as poly (2, 6-dimethyl- 1,4- phenylene ether), poly (2- methyl- 6- ethyl- 1, 4- phenylene ether), poly(2- methyl- 6- phenyl- 1,4- phenylene ether), and the polymers with 2, 6- dimethyl phenol and other phenol groups (for instance, 2, 3, 6- trimethyl phenol and 2- methyl- 6- butyl phenol) can be cited.

(0020)

Within these, a polymer of poly (2, 6- dimethyl- 1,4- phenylene ether), 2, 6- dimethyl phenol and 2, 3, 6- trimethyl phenol is preferable and moreover, poly(2, 6- dimethyl- 1, 4- phenylene ether) is preferable. The PPE production method can be used as long as it is a well known method. For example, the easy production by means of oxidation polymerization of 2,6- xylenol by using a cuprous salt and an amine complex as catalyst in the American patent Number 3306874 according to Hay, and besides this, the production methods that are detailed in the American Patent Number 3306875, similarly Number 3257357 and similarly Number 3257358, patent number S52 - 17880 and patent application S50 - 51197 and similarly Number 63 - 152628.

(0021)

Besides, the PPE that is used in the current invention, other than the PPE mentioned above, it makes nuclear PPE and stearine related monomers or its derivatives to react in the presence of a radical emitting agent and in the absence of which the solution form, solvent form, slurry form are made to react under the temperature 80~350 ° C, as a result of which there is denaturation (the nuclear stearine related monomers and its derivatives are grafted or added at 0.01~10% of part weight) and can be PPE or can be the mixture of the above mentioned PPE and the nuclear denatured PPE in a ratio. The PPE used in the current invention, other than the above mentioned PPE, can be used within the range of 400 w/t parts of high impact poly stearine, that it does not exceed and syngotactic poly stearine, polystearine with regard to 100 w/t parts of PPE 100.

(0022)

Further, the hydrogenated block polymer that can be used as the (c) component of the current invention acts as the impact resistance additive agent and additive of the poly propylene related resin of (a) component and poly phenylene related resin of (b) component, and the block polymer that is made up of at least one polymer block A with the main part as the vinyl aromatic compound, and at least one polymer block B that has the main part as the conjugated diene compound with the total amount as 2~90 % for the 1, 2- vinyl bonds and the 3, 4- vinyl bonds of the conjugated diene compound, to which is then added more than 50 % of hydrogen thus forming the hydrogenated copolymer.

(0023)

For example, it is a hydrogenated compound with a block polymer of vinyl aromatic compound - conjugated diene compound containing the structure A - B, A - B - A, B - A - B - A, (A - B -)₄ - Si, A - B - A - B - A etc. The hydrogenated block polymer that is the (c) component, contains 20 ~95 % of part weight of vinyl aromatic compound with a block polymer before having been hydrogenated, and preferably should have 30~80 % of part weight.

Moreover in the block structure, the polymer block A that has the main part as vinyl aromatic compound and the homopolymer block of the vinyl aromatic compound consists of the structure of the copolymer block made up of a conjugated diene compound and a vinyl aromatic compound that has above 50 % of part weight of vinyl aromatic compound and is preferably more than 70 % of part weight, besides in the polymer block containing the main part as the conjugated diene compound, and the homopolymer block consists of the structure of the copolymer block made up of a conjugated diene compound that has above 50 % of part weight of the conjugated diene compound and is preferably more than 70 % of part weight, and a vinyl aromatic compound.

(0024)

The polymer block A that has the main part of these aromatic compounds, the polymer block B that has the main part as the conjugated diene compound, and the distribution of the vinyl aromatic compounds and the conjugated diene compounds within the molecular chain in the respective polymer blocks, is random and tapered (the monomer components along the molecular chain are increased or decreased), a part of it may be in block form and these can also be easily formed and in the case of the polymer block A having the main part as the nuclear vinyl aromatic compound and the polymer block B having the main part as the nuclear conjugated diene compound, both respectively having more than 2, then both the polymers can respectively have a similar structure or a different structure.

(0025)

As the vinyl aromatic group compounds that make up the structure of the block copolymers, for example, are stearine, α - methyl stearine, vinyl toluene, p- tert- butyl stearine, diphenylene ether of which either one type or two types can be selected but stearine is preferable. Besides, as conjugated diene compounds, for example are buta diene, isopren, 1, 3- pentadiene, 2, 3- dimethyl- 1,3- butadiene, etc of which either one type or two types can be selected but buta diene, isopren and its mixture is preferable.

(0026)

The polymer block that has the conjugated diene compound as the main part, in the micro structure that is present in the block (bond form of the conjugated diene compound) the total amount of the 1, 2- vinyl bonds and the 3, 4- vinyl bonds (abbreviated below as vinyl bond amounts), from the view point of the mixing property, is 2~ 90 %, preferably 2~ 80 %. For example in the polymer block containing buta diene as the main part, the vinyl bond amounts are of 30 ~90 %.

Besides, in the polymer block containing iso pren as the main part, the vinyl bond amounts are of 2 ~ 80 %.The bond form of these conjugated diene compounds are usually found out by using infra red spectrometers and NMR etc.

(0027)

And the mean molecular weight of the block copolymer that contains the above mentioned structure is 5,000~ 1,000,000, preferably 10,000 ~800,000 and more preferably in the range of 30,000 ~ 500,000, and the molecular weight distribution(the ratio between the weight- average molecular weight (Mw) measured by gel permeation chromatography and the number- average molecular weight (Mn)) is less than 10.Moreover, the molecular structure of the block copolymer can be easily mixed with either a linear form, radiative form and branch form.

(0028)

The block polymer that has such a structure, can be used as the current invention's (c) component by using the hydrogenated copolymer(hydrogen is added) to which a lipid group double bond containing polymer block B is added, which has the conjugated diene compound of the above mentioned block copolymer as the main part. The hydrogenation rate of the lipid group double bond is more than 50 %, and should be preferably more than 80 %, the most preferable is more than 90 %. This hydrogenation rate is normally found out by using infra red spectrometer and NMR etc.

(0029)

The hydrogenated block polymers that are the above mentioned (c) component, when containing the above mentioned structure, the production method does not matter. As examples of widely known production methods, they are the production methods that have been cited in Patent Number S46 - 11486, Patent Number S49 - 66743, Patent Number S50 - 75652, Patent Number S54 - 126255, Patent Number S56 - 10542, Patent Number S56 - 62847, Patent Number S56 - 100840, Patent Number H2 - 300318, English Patent Number 1130770 and American patent Number 3281383 and similarly the method in Number 3639517 and the English patent Number 1020720 as well as the American patent Number 3333024 and similarly the production method as cited in number 4501857.

(0030)

The compounds that are used as the(d) component in the current invention and which contain active groups within the molecule that can be selected from carbon- carbon double bonds and carboxylic groups, anhydrous groups, epoxy groups, for which examples have been given and explained. As a compound that contains unsaturated

dicarboxylic groups and anhydrous groups, anhydrous compounds and maleic acid, fumaric acid, chloro maleic acid, cis- 4- cyclohexene- 1, 2- dicarboxylic acid can be cited.

(0031)

Especially α , β - unsaturated dicarboxylic acid and its derivatives, particularly fumaric acid, maleic acid and anhydrous maleic acid is good, but anhydrous maleic acid is the most preferable. The carboxylic groups of these unsaturated dicarboxylic acids have been changed into esters, and at processing temperature, the esters become degraded and the change into compounds containing carboxylic groups becomes possible. As compounds containing epoxy groups, glycidyl metacrylate, allyl glycidyl ether, epoxy natural lipids, etc can be cited.

(0032)

As a chemical compound that contains an amino group, the following can be cited- unsaturated alcohols with the general formula of $C_nH_{2n-3}O$ (n is a positive integer) such as allyl alcohol, 4- penten- 1- ol, 1, 4- penta diene- 3- ol, unsaturated alcohols with the general formula of $C_nH_{2n-5}O$ (n is a positive integer). As an example for a compound containing amino group, the unsaturated alcohol that is the above mentioned unsaturated alcohol with OH group replaced by the NH_2 group can be cited. The chemical compounds cited over here can be used either singly or can be used by mixing more than two types together.

(0033)

Furthermore there is no restriction on the form of the inorganic filler that is used as the (e) component of the current invention and the widely known inorganic fillers can be used. Preferably, it should be of fibre form and / or plate form and can be used by mixing more than two types. Besides, in the surface processing meant for the (d) component used in the current invention, it is preferable to use a processing that uses coupling agents such as cyran related and titanate related, etc varieties.

(0034)

As an example for the inorganic filler of fibrous form, whisker types such as glass fibres, carbon fibres, potassium titanate, valastonite etc., can be cited. From the view point of the effect of the inorganic fillers in fibrous form, L / D that denotes the ratio between the mean diameter of the fibres (d) and the mean length (L), should be preferably more than 5. Subsequently, the examples for inorganic filler of plate form include glass flakes, mica, talc etc. These inorganic fillers of plate form from the view point of the reinforcement effect, R / H that denotes the ratio between mean thickness(H) and mean plate radius(R), should be preferably more than 5.

(0035)

The resin compound of the current invention is a structure that has the basic components of the above mentioned (a) component ~ (e) component. In the current invention, the amount of (a) component is 45 ~ 95 % of part weight from the view point of the flow property, solvent resistance property and the heat resistance property. Subsequently, the amount of (b) component is 55 ~ 5 % of part weight from the view point of the flow

property, solvent resistance property and the heat resistance property. Following this, the amount of (c) component, with regard to (a) component, (b) component 100 w/t parts, is 1 ~30w/t parts from the view point of mechanical strength, impact resistance properties, rigidity property, solvent resistance property and the heat resistance property.

(0036)

Subsequently, the mixing amount of (d) component is 0.1~ 5 w/t parts and preferably 0.1~3 w/t parts from the aspect of emergence of unsoluble compound and the impact resistance properties and flow properties with regard to 100 w/t parts of the total amount of the above mentioned (a) component and the (b) component. Following this, the mixing amount of (e) component is 3~ 100 w/t parts and preferably should be selected within the range of 3~80 w/t parts from the aspect of mechanical strength, rigidity properties, the impact resistance properties and flow properties with regard to 100 w/t parts of the total amount of the above mentioned (a) component ~ (d) component.

(0037)

In the current invention, besides the above mentioned components and in the scope of not losing the effect or the properties of the current invention, the necessary additive components such as the following can be added:- for example, oxidation preventive agents, metallic inactivation agents, flame retardant agents (organic ester related phosphate compounds, ammonium related poly phosphate, aromatic halogen related flame retardants, silicon related flame retardant agents etc), fluorine related polymers, heat plasticizing agents, (low molecular weight poly ethylene), epoxynated soybean oil, poly ethylene glycol, lipid group esters etc), flame retardant supporting agents such as antimony trioxide etc., improvement agents for light resistance properties, agents for poly olefin, slipping agents, inorganic and organic fitting agents and strengthening materials (poly acrylonitrile fibres, carbon black, titanium oxide, calcium carbonate, conductive metal fibres, conductive carbon black etc), each type of color additives, separating agents, etc.

(0038)

The production method of the current invention does not especially have any restrictions and the production can be carried out by using the kneading extractor and the dissolving and kneading machine, and the dissolving and kneading machines that carry out this method are for example, the polyaxial extractors that include the monoaxial extractor, the biaxial extractor, roll, kneader, Brabender -plastograph, Bunbury mixer, etc can be cited as thermal dissolving and kneading machines, among which the dissolving and kneading method is most preferable when employed with biaxial extractor. Examples include the WERNER & PFLEIDERER company made ZSK series, Toshiba Machines company made TEM series, Nippon Seikou company made TEX series, etc.

(0039)

The preferable conditions for the extractor used in the current invention are listed below. The extractor's L / D (barrel effective length / barrel internal diameter) is in the range of above 20 and below 60, and preferably in the range of above 30 and below 50. With regard to the flow of direction of the raw materials in the extractor, there is a first raw

materials supply port and then there is a first vacuum bend in the lower flow region and below this lower flow region, a second raw materials supply port is set up and it is preferable to set up a second vacuum bend in that lower flow region. Within this, a kneading section is set up in the upper flow region of the first vacuum bend, a kneading section is set up between the first vacuum bend and the second raw materials supply port, and it is preferable to set up a kneading section between the second raw materials supply port and the second vacuum bend. The raw materials supply method to the second supply port does not have any limits as such and, when compared to the addition supply from the extractor's second supply release port, it is preferable and the supply is stable when the forcible side feeder is used from the extractor side release port.

(0040)

When the granules, filler, etc are present, the forcible side feeder that is supplied from the side of the extractor, is more preferable, and the upper release port of the second supply port in the extractor can be released in order to let out the air. The temperature of the dissolving and kneading action at this time, is not especially limited to the screw revolution numbers, the normal temperature during the dissolving and kneading is 200~370 ° C, and the screw revolution number can be easily selected from 100 ~1200 rpm.

(0041)

The most preferable production method of the current invention would be to dissolve and knead the entire amount or a part of the (c) component, and the above- mentioned (b) component, the entire amount of the (d) component and the (a) component, following which the dissolving and kneading action is continuously performed after the supply of (a) component, the remaining amount of (c) component and (e) component. By adopting this production method, the resin compound can be obtained with (b) component present in (a) component, mixture of (c) component and (d) component and (e) component, each of which is present in an excellent homogeneously dispersed form, moreover the (d) component's additive effect is present in a remarkable fashion and the (e) component can be controlled to the least limit by the crushing during the dissolving and kneading action, and a resin compound with excellent flow properties and impact resistance properties is obtained.

(0042)

From the first raw material supply port of the biaxial extractor, with regard to the 45~95 % of part weight for the total amount or a part of (b) component, (d) component and (a) component, preferably 5~100 % of part weight of the total (a) component mixed amount and the total 100 w/t parts of the (a) component, (b) component, total amount or part of 1~30 w/t parts of (c) component mixture amount, preferably the (c) component mixture amount was supplied at 30~100 % of part weight and these (a) component ~ (d) component under the conditions of dissolving and kneading, from the second raw materials supply port, the (a) component, the remaining amount of the (c) component and the total amount of the (e) component were supplied by using the forcible side feeder and further more, by conducting the dissolving and kneading actions repeatedly, the resin compound can be obtained with (b) component present in (a) component, mixture of (c) component and (d) component and (e)

component, each of which is present in an excellent homogeneously dispersed form, moreover the (d) component's additive effect is present in a remarkable fashion and the (e) component can be controlled to the least limit by the crushing during dissolving and kneading. This type of production is extremely important in order to obtain a resin compound with excellent flow properties as well as impact resistance properties at the same time.

(0043)

The resin compound of the current invention that is obtained in such a manner is formed by various methods that have been known since before, for example injection molding, extraction molding, extraction variant molding, molding of each type of component due to midair molding. As each type of component, the automobile parts can be given and for instance, external items such as bumper, fender, door panel, each type of mall, emblem, engine hood, foil cap, roof, spoiler, each type of aero parts, etc, internal goods such as instrument panel,, console box, trim etc. and moreover it applies to the secondary battery electric battery components that are equipped in the electric automobiles and hybrid electric automobiles, etc.

(0044)

It can also be used as an internal or external components in electric devices and is used in each type of computer and its peripheral devices, other OA devices, television, video devices, cabinets of each type of disc players, etc, chassis, refrigerators and components used for each type of pump casing etc for components used in industrial use.

(0045)

(Form of implementation of the invention)

About the practical examples of the current invention, it is explained in greater detail and it is not limited to these practical examples.

1. The poly propylene homo poly propylene that is used as the (a) component in the practical example is taken as (a-1) component at solubility point= 169 ° C, MFR = 16. The homo poly propylene that is used as the above- mentioned (a-1) component, is mixed with anhydrous maleic acid(Nippon Yuushi Products company make, crystal MAN) and 2, 5- dimethyl- 2, 5- di (t- butyl p- oxy) hexene in the respective ratios of 100 w/ t parts, 1 w/t part, 1 w/ t part, and under the conditions of using a biaxial extractor at the barrel temperature of 210 ° C, it was extracted and made into a pellet. This was taken as the (a-2) component. The homo poly propylene that was used as the above- mentioned (a-1) component, is mixed with sample glycidyl methacrylate of super- high grade, 2, 5- dimethyl- 2, 5- di (t- butyl p- oxy) hexene in the respective ratios of 100 w/ t parts, 4 w/t part, 1 w/ t part, and under the conditions of using a biaxial extractor at the barrel temperature of 210 ° C, it was extracted and made into a pellet. This was taken as the (a-3) component. The MFR (melt flow rate) of poly propylene is based on ASTM D-1238, and was measured at 230 ° C at load 21.2 N.

(0046)

2. The poly propylene ether of reduced degree of viscosity 0.43 that is obtained by the oxidation polymerization of PPE 2,6- xyleneol which is the (b) component of the

practical example and was taken as (b - 1) component. The above- mentioned (b - 1) component, is mixed with anhydrous maleic acid (Nippon Yuushi Products company make, crystal MAN) and 2, 5- dimethyl- 2, 5- di (t- butyl p- oxy) hexene in the respective ratios of 100 w/ t parts, 1 w/t part, 1 w/ t part, and under the conditions of using a biaxial extractor at the barrel temperature of 300 ° C, it was extracted and made into a pellet. This was taken as the (b - 2) component. The above- mentioned (b - 1) component, is mixed with stearine-maleic anhydride copolymer (Amo chemical company make, dilac 232) in the respective ratios of 97 w/ t parts, 3 w/t part and under the conditions of using a biaxial extractor at the barrel temperature of 300 ° C, it was extracted and made into a pellet. This was taken as the (b - 2) component.

(0047)

3. The hydrogenated block copolymer that is used as (c) component in the practical example has the structure of poly stearine- hydrogenated poly butadiene- poly stearine, is the mixture of a hydrogenated block polymer with the hydrogenation rate to be 99.9 % for the poly buta diene part, 36 % for the total of 1, 2- vinyl bonds for the poly buta diene part before the addition of hydrogen at stearine bonds 67 %, mean molecular weight 42,000 molecular amount distribution 1.05, and is taken to be (c - 1). Similarly, the compound that has the structure of poly stearine - hydrogenated poly butadiene - poly stearine, is the mixture of a hydrogenated block polymer with the hydrogenation rate to be 99.9 % for the poly buta diene part, 50 % for the total of 1, 2- vinyl bonds for the poly buta diene part before the addition of hydrogen at stearine bonds 43 %, mean molecular weight 95,000 molecular amount distribution 1.04, is taken to be (c- 2).

(0048)

Moreover, the polymer (c - 3) was taken to be the mixture of a hydrogenated block polymer with the hydrogenation rate to be 85.4 % for the poly isopren part, 55 % for the total of 1, 2-vinyl bonds and 3, 4- vinyl bonds for poly isopren before the addition of hydrogen at stearine bonds 43%, mean molecular weight 81,000 molecular amount distribution 1.15, and having the structure of poly stearine- hydrogenated poly isopren- polystearine. Moreover, the polymer (c - 4) was taken to be the mixture of a hydrogenated block polymer with the hydrogenation rate to be 99.3 % for the poly isopren part, 4 % for the total of 1, 2-vinyl bonds and 3, 4- vinyl bonds for poly isopren before the addition of hydrogen at stearine bonds 65 %, mean molecular weight 91,000 molecular amount distribution 1.13, and having the structure of poly stearine- hydrogenated poly isopren- polystearine.

(0049)

The above mentioned (c - 10) component, was mixed in the ratio of 100 w/t parts, 1w/t part, 1w/t part respectively for anhydrous maleic acid (Nippon Yuushi Products company make, Crystal MAN) and 2,5- methyl- 2,5- di (t- butyl p-oxy) hexene,

extracted under the conditions of barrel temperature 230 ° C by using a biaxial extractor and was made into a pellet. This was taken as the (c - 5) component.

(0050)

4. Within the (d) molecule used in the practical example, a compound containing the active groups that can be selected from carbon- carbon double bonds and carboxylic acid groups, anhydride groups, epoxy groups, amino groups and hydroxy groups.

The anhydrous maleic acid (Nippon Yushi Products Company make, Crystal MAN) was taken as (d - 1). The sample glycyd metacrylate of super high grade was taken as (d - 2).

(0051)

5. The (e) inorganic filler used in the practical example has a mean diameter 13 μ , and is surface- processed with an amino cyan related coupling agent, and glass fibres were taken as (e - 1). The glass flakes with mean diameter 600 μ , and is surface- processed with an amino cyan related coupling agent, were taken as (e - 1).

(0052)

(Practical examples 1~17 and comparative examples 1~13)

By using a biaxial extractor ZSK-40 (WERNER & PFLEIDERER Company make), with regard to the direction of flow of materials, a first materials supply port and in the lower current a second materials supply port was set up and furthermore, a vacuum bend was set up in the lower current. The raw materials supply method to the second supply entry was by using a forcible side feeder from the extractor side release port. By using the above- mentioned extractor as set up, (a) poly propylene, (b) poly phenylene ether, (c) hydrogenated block copolymer used as additive, (d) unsaturated compound containing active groups, (e) inorganic filler were mixed and composed as shown in the table 1, under the conditions of the extraction temperature at 280~300°C, the screw revolution number at 200rpm, discharge amount at 80 kg/ hour and by dissolving and kneading, a pellet was obtained. The resin compound pellet thus obtained is used to measure the melt flow rate (MFR) (250 ° C, load 98N).

(0053)

Further, by using the resin pellet obtained as mentioned above, and supplying it into a screw incline type injection molding machine set at 240 ~280 ° C and under the conditions of tool temperature 60 ° C, the test piece used for the izode impact test, the test piece used for the curvature elasticity modulus measurement test and the test piece used for the measurement of the deflection temperature under load are molded by injection molding, and by using gear open and leaving for 46 hours under a 80 ° C environment ,the thermal history processing was then carried out.

(0054)

Subsequently, these test pieces were used and the izode (notch attached) impact strength (ASTM D-256: 23 ° C), the curvature elasticity modulus (ASTM D - 790) and the deflection temperature under load(ASTM D - 648) were measured. Furthermore, the fracture surface of the test piece when the izode impact strength measurement was carried

out with FE- SEM/ JSM- 6700F (Nippon Electric Company make) and observed at 350 times magnification, and the attachment of the resin to the protruding inorganic filler, was observed. This observation result is given as “ O” when one piece of the inorganic filler definitely has the resin attached to one part, “Δ’ when there is a mixture of resin that is not visibly attached to the inorganic filler, and as “X” when there is no attachment of the resin at all.

(0055)

The above results were depicted in table 1~ table 4. From table - 1, the resin compound of the current invention does not bear any relation to the type of inorganic filler, and has excellent impact resistance properties as well as flow properties at the same time, and it was evident that it is not possible to simultaneously improve the impact resistance properties as well as the flow properties when the composition of the current invention falls outside the scope.

(0056)

(Table – 1)

[illegible]

Second raw materials supply port	(a) Component (a-1)	% of part weight	50	50	50	50	50	0	60	50
	(a) Component (a-2)	% of part weight								
	(a) Component (a-3)	% of part weight								
	(b) Component (b-1)	% of part weight								40
	(b) Component (b-2)	% of part weight								
	(b) Component (b-3)	% of part weight								
	(c) Component (c-1)	W/t parts						5		
	(c) Component (c-2)	W/t parts								
	(c) Component (c-3)	W/t parts								
	(c) Component (c-4)	W/t parts								
	(c) Component (c-5)	W/t parts								
	(d) Component (d-1)	W/t parts								
	(d) Component (d-2)	W/t parts								
	(e) Component (e-1)	W/t parts	50	50	50	50	50	50	50	50
	(e) Component (e-2)	W/t parts								
Physical properties	Melt flow rate	g/10min	21	20	22	23	35	21	18	16
	Izode impact strength	J/m	170	180	190	150	90	160	140	130
	Curvature elasticity modulus	Mpa	7210	7180	7230	7440	6660	7100	6820	6430
	Deflection temperature under load	°C	158	155	156	161	143	156	154	154
Attachment of resin to inorganic filler			0	0	0	0	0	0	0	Δ

(0057)
(Table-2)

		Practical example 11	Practical example 12	Practical example 13	Practical example 14	Practical example 15	Practical example 16	Practical example 17
First raw materials supply port	(a) Component (a-1)	10	10	10	10	60		
	(a) Component (a-2)							
	(a) Component (a-3)							
	(b) Component (b-1)	40	40	40	40		40	
	(b) Component (b-2)							
	(b) Component (b-3)							
	(c) Component (c-1)	0	10	10	10			10
	(c) Component (c-2)							
	(c) Component (c-3)							
	(c) Component (c-4)							
	(c) Component (c-5)							
	(d) Component (d-1)	0.5	0	0.5	0.5	0.5	0.5	0.5
	(d) Component (d-2)							
	(e) Component (e-1)			50	25			
Second raw materials supply port	(e) Component (e-2)							
	(a) Component (a-1)	50	50		50		60	60
	(a) Component (a-2)							
	(a) Component (a-3)							
	(b) Component (b-1)					40		40
	(b) Component (b-2)							

[illegible]

(0056)
(Table - 3)

		Compar ative exempl e 1	Compar ative exempl e 2	Compar ative exempl e 3	Compar ative exempl e 4	Compar ative exempl e 5	Compar ative exempl e 6	Compar ative exempl e 7	Compar ative exempl e 8	Compar ative exempl e 9	Compar ative exempl e 10
	(a) Component (a-1)	10	10	10	10	10	10	10	10		
	(a) Component (a-2)	% of part weight								10	
	(a) Component (a-3)	% of part weight									10
	(b) Component (b-1)	% of part weight	3	70	40	40	40	40	40	40	40
	(b) Component (b-2)	% of part weight									
	(b) Component (b-3)	% of part weight									
	(c) Component (c-1)	W/t parts	10	10	40	10	10	10	10	10	10
	(c) Component (c-2)	W/t parts									
	(c) Component (c-3)	W/t parts									
	(c) Component (c-4)	W/t parts									
	(c) Component (c-5)	W/t parts									
	(d) Component (d-1)	W/t parts	0.5	0.5	0	0	0.5	0.5			
	(d) Component (d-2)	W/t parts									
	(e) Component (e-1)	W/t parts									
	(e) Component (e-2)	W/t parts									
	(a) Component (a-1)	% of part weight	87	20	50	60	50	60	50		
	(a) Component (a-2)	% of part weight								50	
	(a) Component (a-3)	% of part weight									50
	(b) Component (b-1)	% of part weight									
	(b) Component (b-2)	% of part weight									

First raw
materials supply
port

Second raw
materials supply
port

[illegible]

(0059
(Table-4)

			Comparative example 11	Comparative example 12	Comparative example 13
First raw materials supply port	(a) Component (a-1)	% of part weight	10	10	10
	(a) Component (a-2)	% of part weight			
	(a) Component (a-3)	% of part weight			
	(b) Component (b-1)	% of part weight			40
	(b) Component (b-2)	% of part weight	40		
	(b) Component (b-3)	% of part weight		40	
	(c) Component (c-1)	W/t parts	10	10	
	(c) Component (c-2)	W/t parts			
	(c) Component (c-3)	W/t parts			
	(c) Component (c-4)	W/t parts			
	(c) Component (c-5)	W/t parts			10
	(d) Component (d-1)	W/t parts			
	(d) Component (d-2)	W/t parts			
	(e) Component (e-1)	W/t parts			
	(e) Component (e-2)	W/t parts			
Second raw materials supply port	(a) Component (a-1)	% of part weight	50	50	50
	(a) Component (a-2)	% of part weight			

	(a) Component (a-3)	% of part weight			
	(b) Component (b-1)	% of part weight			
	(b) Component (b-2)	% of part weight			
	(b) Component (b-3)	% of part weight			
	(c) Component (c-1)	W/t parts			
	(c) Component (c-2)	W/t parts			
	(c) Component (c-3)	W/t parts			
	(c) Component (c-4)	W/t parts			
	(c) Component (c-5)	W/t parts			
	(d) Component (d-1)	W/t parts			
	(d) Component (d-2)	W/t parts			
	(e) Component (e-1)	W/t parts	50	50	50
	(e) Component (e-2)	W/t parts			
Physical properties	Melt flow rate	g/10min	13	16	14
	Izode impact strength	J/m	110	80	70
	Curvature elasticity modulus	Mpa	5950	5700	5630
	Deflection temperature under load	°C	146	138	141
Attachment of resin to inorganic filler			O	A	A

(0060)

(Effect of invention)

In the resin compound that is made up from poly propylene related resin, the poly phenylene ether related resin, the hydrogenated block copolymer being used as an additive and as an impact resistance additive and the inorganic filler, unsaturated compounds containing specific active groups are used and moreover, by using a specific production method, a resin compound that had excellent impact resistance properties as well as flow properties at the same time, could be obtained.

Continuation of front page

(51) Int.Cl.⁷

Distinguishing symbol

FI

Theme code (reference)

C 08 L

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C 08 J 53 / 02

71 / 12

71 / 12

F term (Reference) 4J002

BB11W

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BP013

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CH07X

DA017

DE137

DJ047

DJ057

DL007

EC036

EF076

EH106

EL036

EL146

EN106

FA007

FA047

FD13

GN00

GQ00
